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Title: R&D on MgB2 at LANL for Applications to Superconducting RF Cavities

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R&D on MgB₂ at LANL for Applications to Superconducting RF Cavities

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Mechanical Design & Engineering Group

Accelerator Operations & Technology Division

A seminar at KEK, 17 February 2020

LA-UR-20-XXXXX





Outline

- Introduction
- Brief history of my research on MgB₂
- Recent activities and results





Introduction

- Recent SRF cavities (especially 1.3 GHz elliptical cells) made of bulk Nb are approaching its theoretical limit of $E_{acc} \sim 50$ MV/m ($B_{peak} \sim 200$ mT).
- While it is important to increase quality factor and production reliability and yield of high-quality Nb SRF cavities, finding a new material that could overcome the limit of Nb technology is becoming increasingly important for SRF technology to be more attractive and to open up other opportunities.
- Nb₃Sn technology has become close to the application level.
- MgB₂ cavities of practical size have not yet been produced yet, but its high T_c of ~40 K is very attractive to enable the operation at a higher temperature such as 20-25 K with cryocoolers.



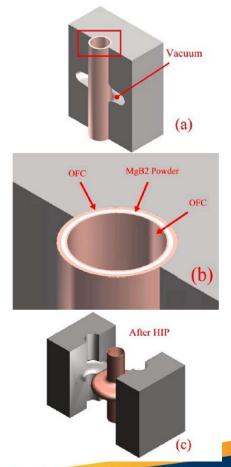


Slide 4

Brief history of my research on MgB₂

- Soon after MgB₂ was discovered to be superconducting at 39 K in 2001, a feature written in a review paper caught my eyes, "absence of weak links" (at the grain boundaries). (These weak links had prevented high-T_c materials from being applied for SRF cavities.)
- My first paper on MgB₂ at EPAC2002 proposed to use thick MgB₂ layer formed on a copper cavity using hot isostatic press (HIP).

At that time, I did not know that bulk or thick film is not so useful.







Slide in 2004, I wrote a paper with Collings and Sumption of OSU. The current cavity coating idea is similar to the idea written in this paper.

INSTITUTE OF PHYSICS PUBLISHING

SUPERCONDUCTOR SCIENCE AND TECHNOLOGY

Supercond. Sci. Technol. 17 (2004) S595-S601

PII: S0953-2048(04)77346-7

Magnesium diboride superconducting RF resonant cavities for high energy particle acceleration

E W Collings¹, M D Sumption¹ and T Tajima²





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² Los Alamos Neutron Science Center, Accelerator Physics and Engineering, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

In 2005, we showed that R_s can be lower than that of Nb and there is little increase in R_s with fields up to 120 Oe limited by available power [Tajima et al. PAC2005]

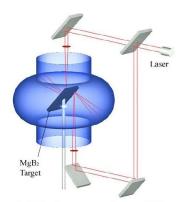


Figure 7: An idea for coating a cavity using a MgB₂ target and a KrF excimer laser.

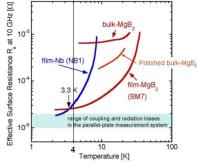


Figure 3: Surface resistance vs. temperature of a 400 nm MgB₂ film coated on a sapphire substrate. Bulk samples and Nb data are shown for comparison. [6]

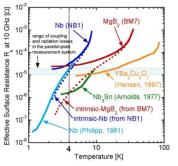


Figure 4: Prediction of intrinsic (BCS) surface resistance (dotted line) from experimental data. [6]

Deposited with reactive co-evaporation at Superconductor Technologies, Inc. (STI)



Figure 5: MgB₂ coated Nb disks of 14.6 mm in diameter.

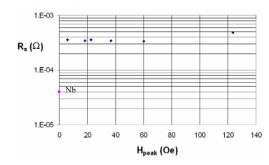


Figure 6: R_s of the Nb TE_{011} mode cavity with a MgB_2 sample at the center of the bottom plate, as a function of the peak magnetic field on the sample. The data was converted to 10 GHz using an f^2 law.

Collaboration with Romanenko at Cornell





In 2005-2006, we proposed to coat cavity with pulsed laser deposition (PLD), but the PLD films had poor quality [Tajima et al. EPAC2006]

 $T_c \sim 27 \text{ K}$

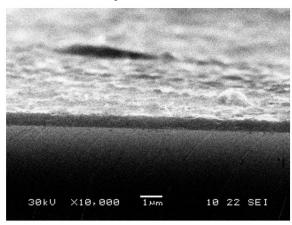


Figure 3: A SEM image of the cross section of off-axis PLD MgB₂ film (ID: 300705v) on Al₂O₃-C substrate. The film thickness is 500-700mm

Tested by Romanenko at Cornell

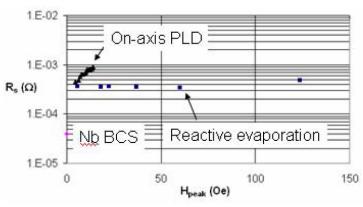


Figure 6: Surface resistance at 10 GHz as a function of surface magnetic field. The data was scaled from 6 GHz data using f^2 law.

Collaboration with Yue Zhao of U. Wollongong, Australia





In 2007, we started high-power RF tests with 2-inch diameter disks in collaboration with SLAC using 11.4 GHz RF short pulse (\sim 1 μ s) and a TE₀₁₃-mode cavity

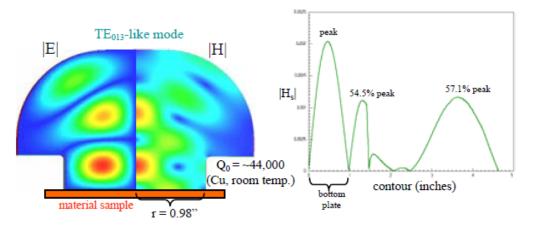


Figure 2: Electric and magnetic fields in the "mushroom" cavity (left) and magnetic field profile along the surface of the cavity (right).

[Tantawi et al. PAC2007]



Slide 9

Due to the low Q of the host cavity made of copper, R_s did not have enough sensitivity. Also, thermal effect seems to have been involved despite a short pulse is used.

STI film deposited at 550 °C on top of ALD alumina

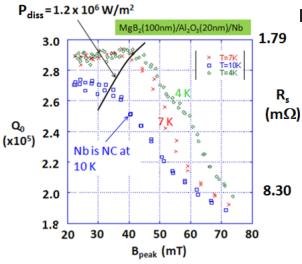


Figure 11: Q_0 as a function of B_{peak} at 4 K, 7 K and 10 K for the $MgB_2(100 \text{ nm})/Al_2O_3(20 \text{ nm})/Nb$ sample shown in Fig. 8. The solid line is a curve described by Eq. (2) with $P_{diss} = 1.2 \times 10^6 \text{ W/m}^2$.

Inter-diffusion layer causes high RF loss

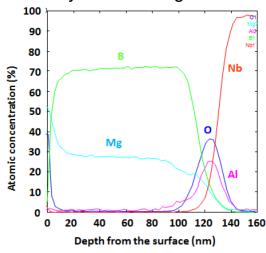


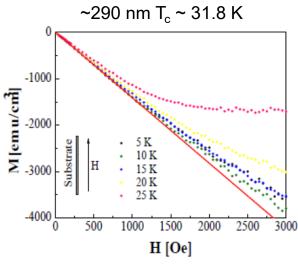
Figure 12: Auger depth profile of the MgB₂(100 nm)/Al₂O₃(20 nm)/Nb sample shown in Fig. 8. Interdiffusion of coating components at the interfaces is observed.

[Tajima et al., SRF2011]





In 2010, we started using magnetization measurements to evaluate vortex penetration field B_{vp} to determine the fundamental limit of MgB₂ thin films



E-beam coevaporation at
250 °C
Base p ~ 1 x 10-9
Torr
Collaboration
with Doi et al. of
Kagoshima U.,
Japan

Figure 4: Magnetization curves as a function of applied magnetic field at various temperatures for ~290 nm thick MgB₂ film ($T_c \sim 31.8$ K) deposited on a Si substrate at Kagoshima Univ.

[Tajima, Haberkorn, Civale et al. LINAC2010]

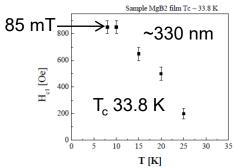


Figure 7: H_{c1} vs. sample temperature from magnetization measurements of ~330 nm thick MgB₂ sample prepared by Kagoshima Univ.

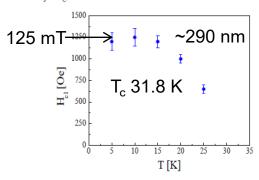


Figure 8: H_{c1} vs. sample temperature from magnetization measurements of ~290 nm thick MgB₂ sample ($T_c \sim 31.8$ K) prepared by Kagoshima Univ.





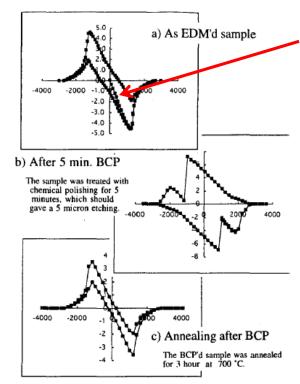


Fig. 2. Hysteresis appeared in magnetization curves. a) as EDM'd sample, b) after chemically polished the EDM'd sample, c) annealed after the CP'd sample.

We are looking at only this part

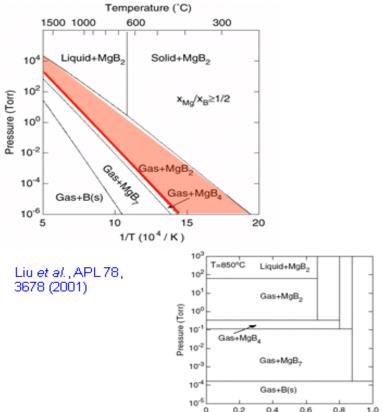
Magnetization measurements were used for SRF cavity research in 1995 by Saito and Wake of KEK to evaluate the effect of surface treatments by looking at hysteresis curves.

The larger hysteresis means more imperfections and/or dislocations since vortices get pinned by them.

[Saito, Wake, SRF1995]



Keys to Growth of MgB₂ Films



Keep a high Mg pressure for phase stability

For example, at 600° C Mg vapor pressure of 0.9 mTorr or Mg flux of 500 Å/s is needed

- No need for composition control, as long as the Mg:B ratio is above 1:2.
- Keep oxygen away: Mg reacts strongly with oxygen - forms MgO, reduces Mg vapor pressure.
- Avoid carbon: Carbon doping reduces T_c and increases resistivity

[Xi, Thin film workshop, JLAB, 18-20 July 2012]





Atomic Fraction of Boron

Summary of MgB₂ deposition techniques that we have evaluated

Reactive Co-evaporation by Brian Moeckly et al., at STI

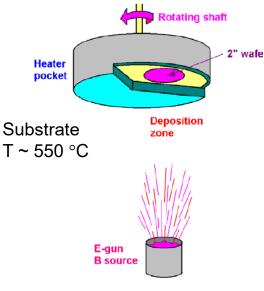


Figure 1: MgB₂ coating system at STI. [5]

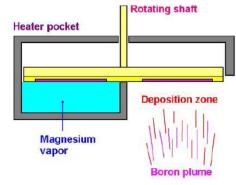


Figure 2: Cross section of the deposition chamber. [5]

[Moeckly et al., ASC2004] [Tajima et al., PAC2005]

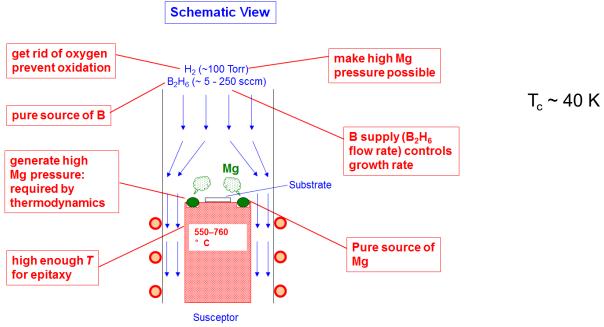


 $T_{\rm C} \sim 39~{\rm K}$



Summary of MgB₂ deposition techniques that we have evaluated (cont.)

Hybrid Physical-Chemical Vapor Deposition



[Xi, Thin film workshop, JLAB, 18-20 July 2012]

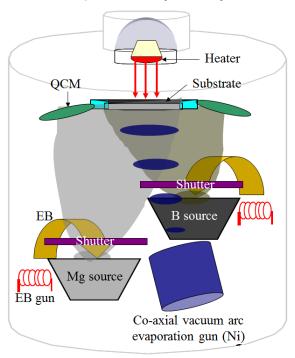


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Summary of MgB₂ deposition techniques that we have evaluated (cont.)

E-beam co-evaporation by Toshiya Doi et al. of Kagoshima U., Japan



Requires base pressure <~1 x 10⁻⁹
Torr to avoid the effect of oxygen

$$T_c \sim 32 \text{ K}$$

Substrate T ~ 250 °C

[Nagatomo et al., Physica C 426 (2005) 1459]





Summary of MgB₂ deposition techniques that we have evaluated (cont.)

Off-axis Pulsed Laser Deposition (PLD) by Y. Zhao of U. Wollongong, Australia

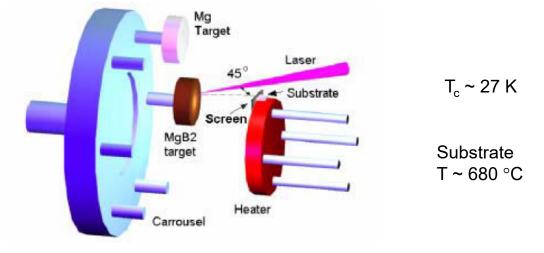


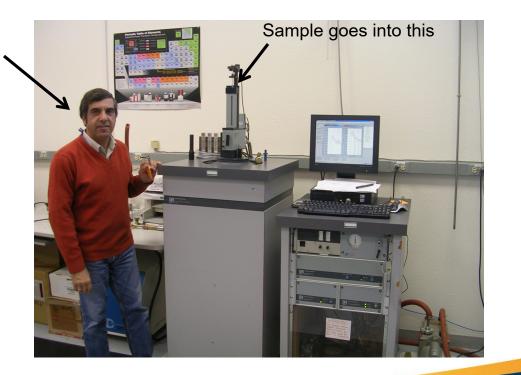
Figure 1: An illustration of off-axis PLD [1].

A KrF laser (λ =248 nm, 25 ns) was used in 120 mTorr Ar atmosphere, then an *in-situ* annealing was carried out at 680 °C for 2 min in a 760 Torr Ar atmosphere [7].



Magnetic vortex penetration field (B_{vp}) measurements using a Quantum Design SQUID magnetometer seem to be a reliable method to determine a fundamental limit

At LANL, Leonardo Civale and his postdocs have been carrying out these measurements



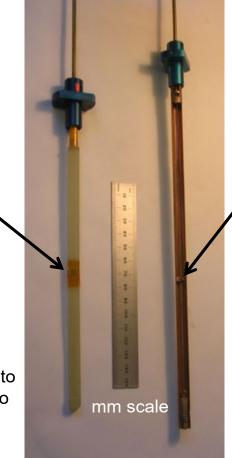


Samples

Typically 6 mm x 6 mm for coated films

Nb reference was a rod with 2 mm diameter and 10 mm long cut out from a single grain RRR>300 Nb sheet

Rod or ellipsoid is better due to less edge effect, but difficult to uniformly coat

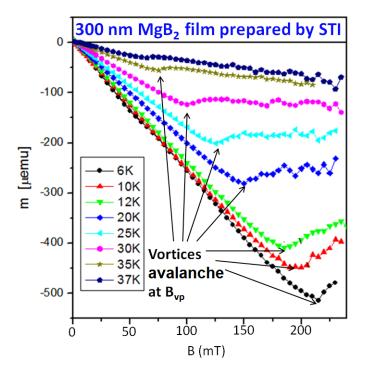


This sample holder has an angle adjustment mechanism

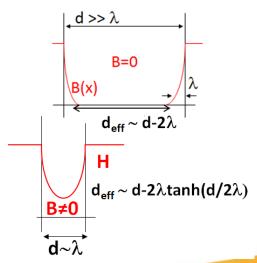




Meissner slope is proportional to the volume of the film, making it difficult to measure ultra-thin films due to small signal. Our present limit is ~200 nm

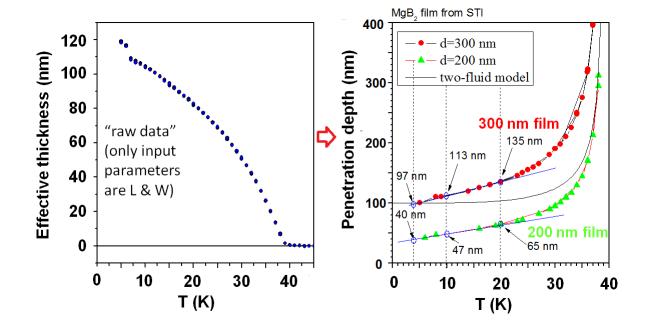


Meissner state: $dm/dH = -V_{eff}/4\pi \propto d_{eff}$ Slope changes with T due to the change in λ





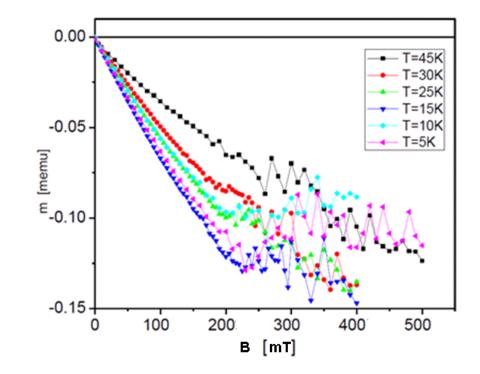
Penetration depth can be estimated from effective thickness. Penetration depth increases with higher temperature. This causes the reduction of effective thickness.





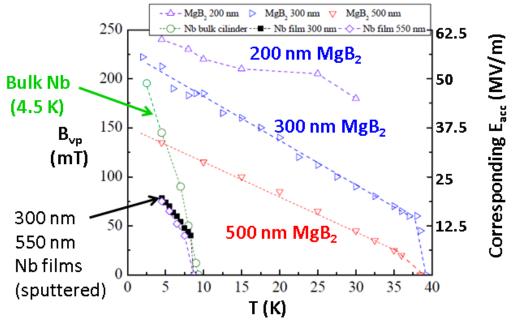


With a 200 nm sample, the signal got quite noisy at high fields, but was still measurable.





Summary of B_{vp} for STI films (200, 300 and 500 nm) compared with cavity-grade bulk Nb and sputtered Nb film. MgB_2 thin films show remarkably high B_{vp} !!



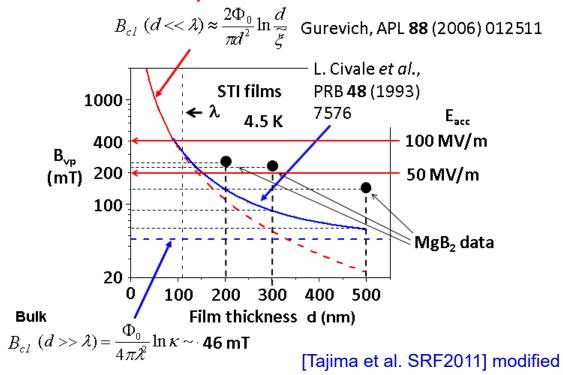
[Tajima et al. SRF2011] modified

Assume $B_{peak}/E_{acc} = 4 \text{ mT/(MV/m)}$



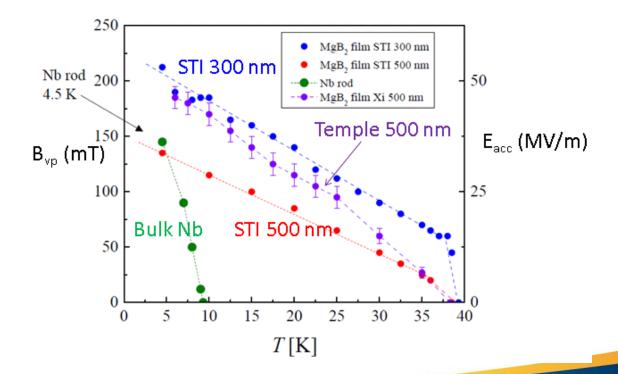


Comparison with theoretical curve of B_{c1} for thin films assuming λ = 110 nm and ξ = 6 nm. Important finding here was that even the films with d> λ have high B_{vp}



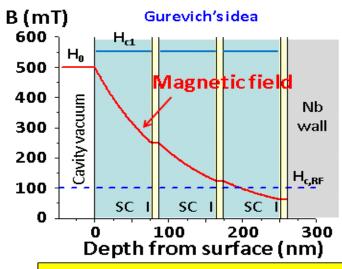


Temple University (Xiaoxing Xi's group) samples prepared with hybrid physical chemical vapor deposition (HPCVD) also showed high $B_{\nu\rho}$



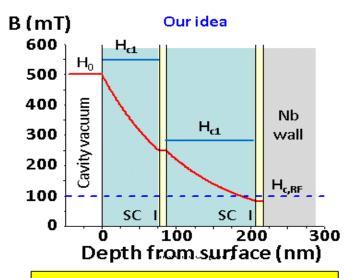


There is a possibility that we can achieve ~500 mT ($E_{\rm acc}$ ~125 MV/m) even with 2 layers of MgB₂





- *d* ≤ 77nm for H_d ≥ 5500 Oe
- 3 layers needed
- coating curved walls with very thin uniform of layers is challenging



Variable thickness multilayers:

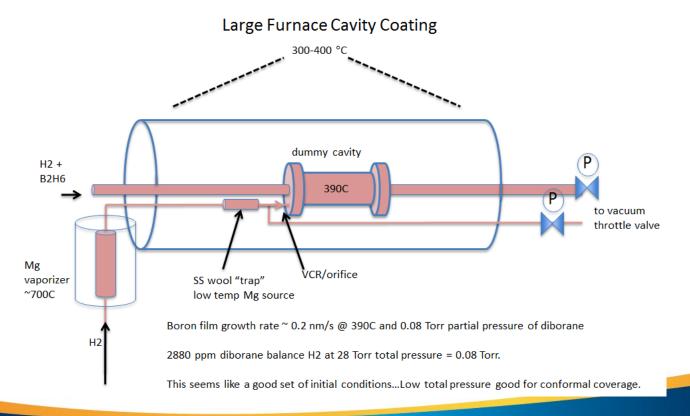
- $d_1 \le 77$ nm for $H_{c1} \ge 5500$ Oe
- only 2 layers needed
- 2nd layer is thicker: 100nm ≤ d₂ ≤
 120nm

[Tajima et al., SRF2011]



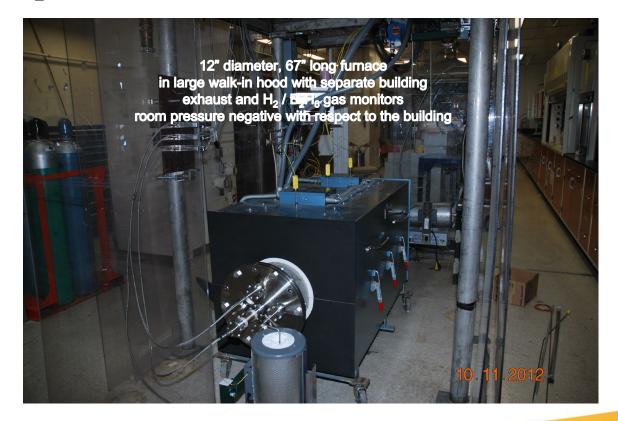


We tried to coat a uniform B layer using diborane (B_2H_6), then react it with Mg vapor to form a uniform MgB₂ layer

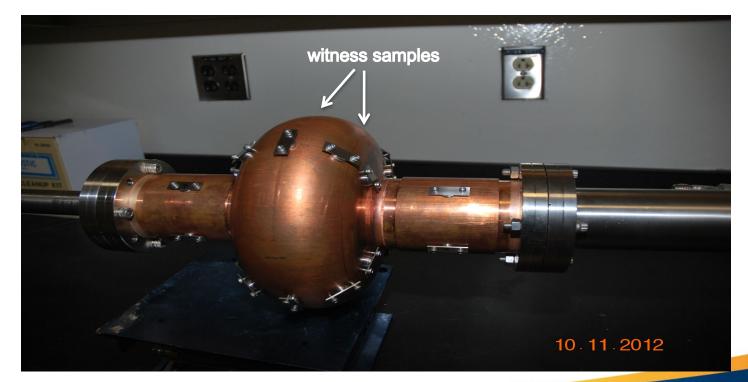




MgB₂ CVD reactor for a 1.3 GHz cavity at TA-35-213



A 1.3 GHz copper cavity with 28 holes for samples. Many samples were 6 mm x 6 mm x 0.43 mm sapphire







The experiments moved to TA-53-0017 (SRF Lab at LANSCE) in 2015 for the following reasons.

- We can accelerate the tests since it is at a non-secure location, i.e., uncleared US citizens and foreign nationals can work without being escorted.
- Close to the infrastructure for SRF cavity preparation and testing (HPR, clean room, cavity testing environment, etc.).
- The TA-35-213 infrastructure was dismantled and the area was changed for other activities.



Results of the tests at TA-53-0017 (SRF Lab) in 2015 (the final year of previous project)

- We focused on the reaction of B with Mg since we did not have to use toxic B₂H₆ gas and we already had many Boron films from the work at TA-35-213.
- We found the conditions to get superconducting MgB₂ films.
 - Confine B film with a Mg pellet and Ar gas and heat it up.
 - The details are in [Tajima et al., SRF2015, p. 700]



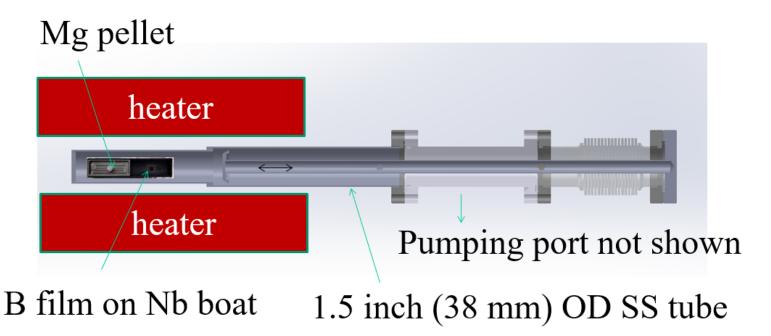
Procedure to get MgB₂ at the tests in 2015

- Bake the system at ~150 °C under vacuum for ~20 min
- Fill the system with UHP Ar gas up to 1/3 psi
- Plug the hot zone with a Mg pellet and a B film
- Raise the temperature to a planned value
- Hold it for 50 min
- Quench it to ~40 °C in ~13 min





Experimental test set up in 2015. The same set up was used for recent tests as well.

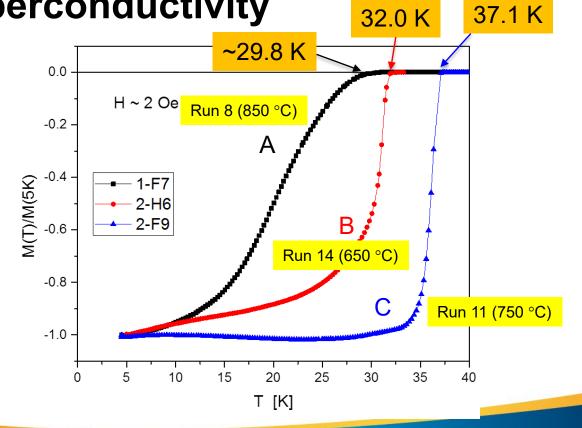




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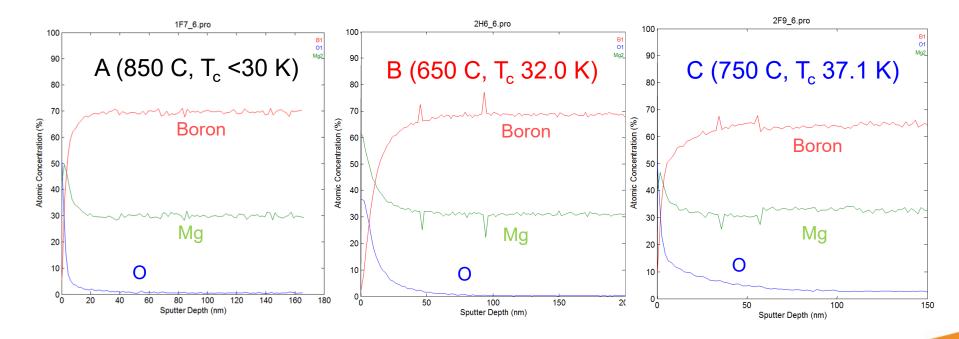


Magnetization measurement results to check the superconductivity





Auger Electron Spectroscopy (AES) depth profile showed the one at 750 C has the closest stoichiometric ratio of Mg:B = 1:2







From 2018, we restarted MgB₂ work as a part of US-Japan Cooperation Project

LANL

- Design and build a new coating system to coat 1.3 GHz 1-cell cavities
- Provide them to KEK for testing with new diagnostics system to be developed.

KEK

- Develop a fixed (and rotating arm) T-map system.
- Develop a magnetic field mapping system.
- Test the MgB₂ cavities provided by LANL.
- Due to insufficient funding, building the new coating system has been delayed.
- We restarted the parameter optimization work using a small existing system.
- The following slides were presented by Sakai-san at the TTC meeting held at CERN on 05 Feb. 2020. They summarize the recent MgB₂ work at LANL.





MgB₂ work at LANL in collaboration with KEK

LANL: Tsuyoshi Tajima, Paolo Pizzol, Anju Poudel, Leonardo Civale, Ivan Nekrashevich, Roland Schulze

KEK: Hiroshi Sakai, Takafumi Okada, Eiji Kako, Kensei Umemori, Taro Konomi





TTC Meeting, CERN, 04-07 February 2020

Outline

- A short description of the coating technique developed at LANL.
- Recent test results
- Design of the coating system to coat 1.3 GHz elliptical cavities.

LANL coating technique

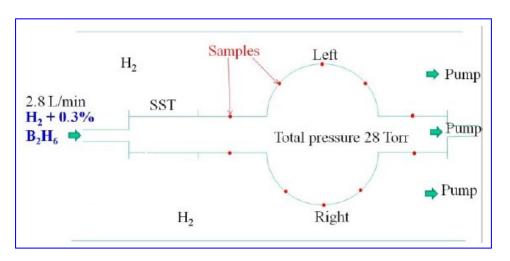
- LANL had a project on MgB₂ from 2010 2015. We developed a technique to coat boron (B) first, then react it with Mg vapor (2-stage process). [e.g. Tajima et al., SRF2015, p. 700]
- We restarted the MgB₂ work as a US-Japan Cooperation Project in 2018.
 - Since we lost the large furnace and equipment at TA-35 that were used in the previous project, they started to design a new coating facility at TA-53 (Tsuyoshi's lab) based on their previous experience at TA-35.
 - In November, 2019, we restarted experiments to optimize parameters for B and Mg reaction using a small system at TA-53 and the B samples obtained in the previous project.

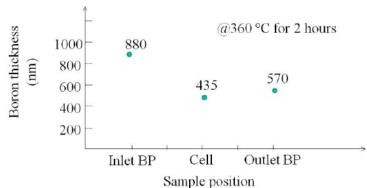
LANL coating technique (1st stage: coating of B layer)

- Flow B₂H₆ gas inside the cavity while keeping the cavity surface at a temperature 250 – 400 °C.
- The B₂H₆ decomposes and a B layer is formed on the cavity surface.
- By controlling the temperature, deposition rate can be controlled, i.e., the higher the temperature, the higher the deposition rate. Thereby the thickness profile can be controlled.
- Usually, cell is thinner than beam pipes.









If the Inlet BP temperature is lower, e.g., 250 °C, the thickness can be reduced.





LANL coating technique (2nd stage: reaction of B layer with Mg vapor)

- Prebake the system up to 200 °C under vacuum.
- Cool down to room temperature, add Mg pellets, bake out the system at 150 °C for >1 h under vacuum.
- Fill the chamber with UHP Ar gas up to 1/3 psi
- Plug the reaction zone to confine Mg vapor
- Heat it up to a planned temperature such as 750 °C and hold it at the temperature for planned period of time.
- Cool down the system fast enough to prevent formed MgB₂ from decomposing.





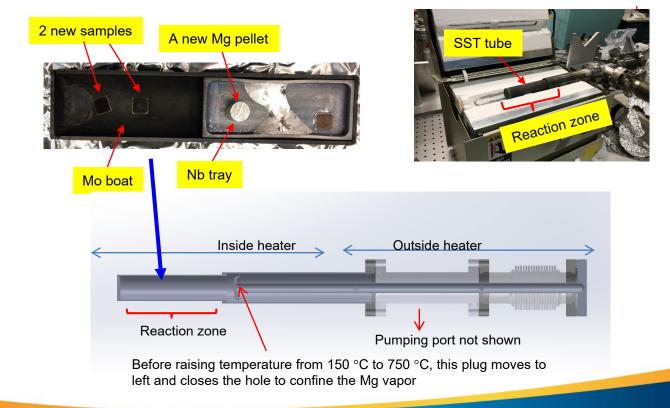
The B films obtained from the previous project

- The previous project used a 1.3 GHz elliptical 1-cell cavity with coupons attached on inlet and outlet beam pipes and cell equator.
- 22 coating runs at TA-35 were performed during the previous project and most of unused samples have been kept in a vacuum desiccator. Most samples have pure B or B with <15 % Mg, 100 – 1000 nm thick.
- They are stored in 3 carriers with each carrier having 81 samples of 6 mm x 6 mm with either sapphire or Nb substrate.
- The sample designation is carrier # column row # such as 2-B4.





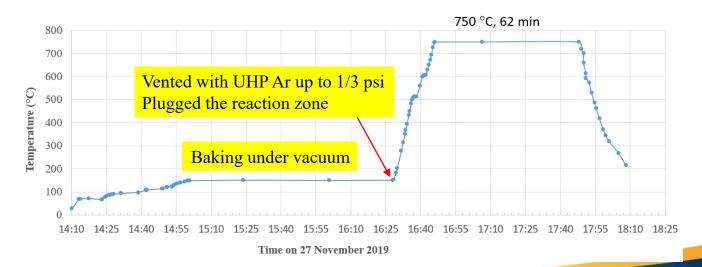
We restarted the test that ended on 28 June 2015 (run 15)





Recent 2 tests to produce MgB₂ film by reacting B film with Mg vapor

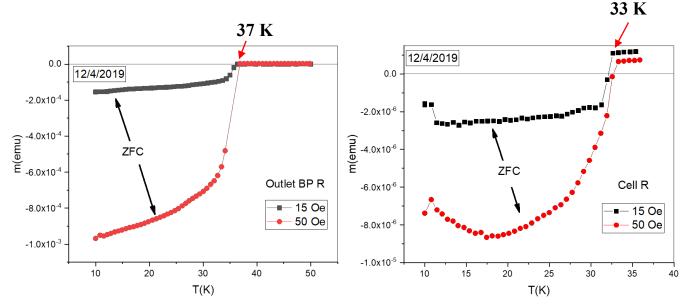
- TA-53 Run 16 on 27 November 2019
 - Hiroshi Sakai and Takafumi Okada from KEK joined LANL workers (Tsuyoshi Tajima and Paolo Pizzol)





Magnetometer measurement to check superconductivity

 This was carried out by Ivan Nekrashevich and Leonardo Civale at TA-3 in Leonardo's lab.

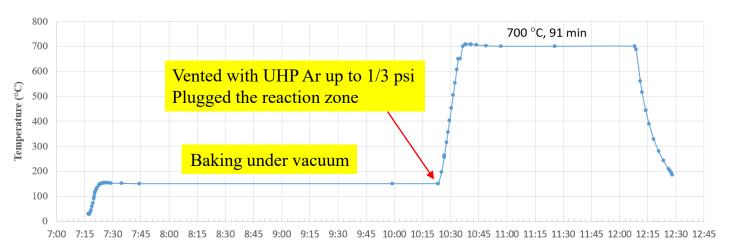






Recent 2 tests to produce MgB₂ film by reacting B film with Mg vapor (cont.)

- TA-53 Run 17 on 20 January 2020
 - Tsuyoshi Tajima did it on his own.

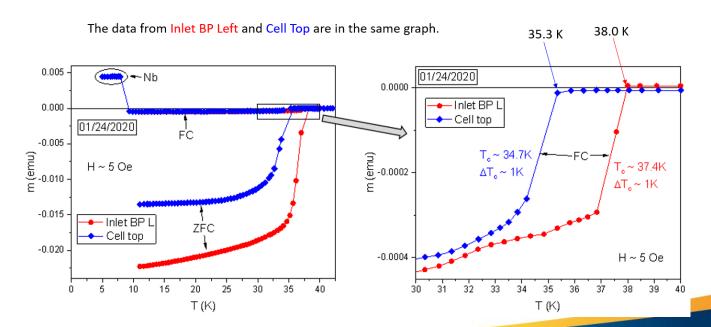


Time on 20 January 2020



Magnetometer measurement to check superconductivity

 This was carried out by Ivan Nekrashevich and Leonardo Civale at TA-3 in Leonardo's lab.





Summary of recent tests and next steps for parameter optimization

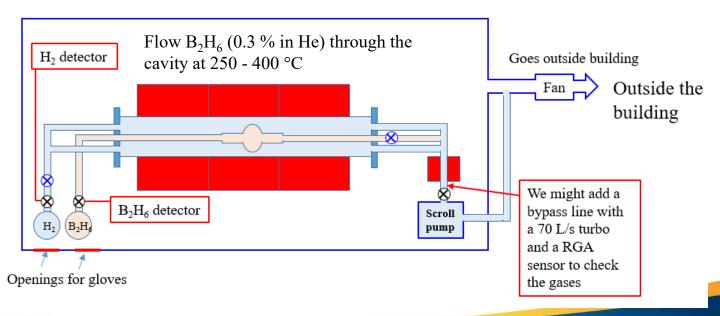
- We were able to reproduce the 750 °C result obtained in 2015.
- The test result at 700 °C was as good as that at 750 °C. (No test at 700 °C in 2015)
- Cell samples showed 3-4 K lower T_c. Need to identify the reason.
- Will test at 650 °C for 1.5 hours to check if we can reproduce the result in 2015 (T_c ~ 32 K at outlet BP top)
 - If T_c is >35 K, try 600 °C test.
 - If T_c is <35 K, try 650 °C for 3 hours to see if T_c goes up with long reaction time.
- Will test longer cooling time at the lowest reaction temperature that gives $T_c > 35$ K.
- Will start sample characterizations using AES/XPS from around April or May.



Design of new coating system (1st stage)

Heater

- All heaters and valves are remotely controlled through Ethernet
- ⊗ Open/close valve (interlocked with gas detectors)
- Pressure control valve including pressure (and flow) sensors

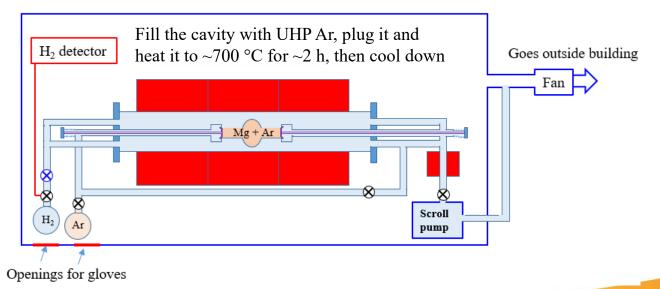




Design of new coating system (2nd stage)

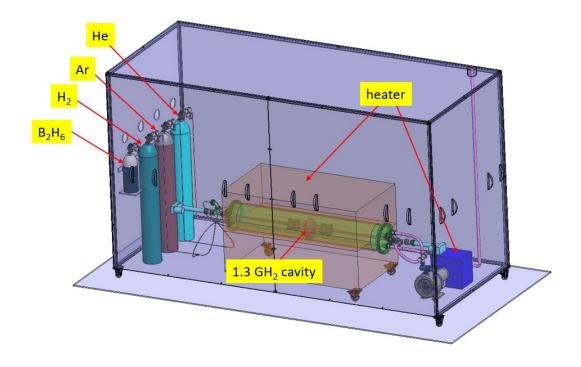
Heater

- All heaters and valves are remotely controlled through Ethernet
- ⊗ Open/close valve (interlocked with gas detectors)
- Pressure control valve including pressure (and flow) sensors





A 3D model has been constructed



The following slides show some new results on the thickness measurements of boron samples obtained in 2013 on 06 February 2020



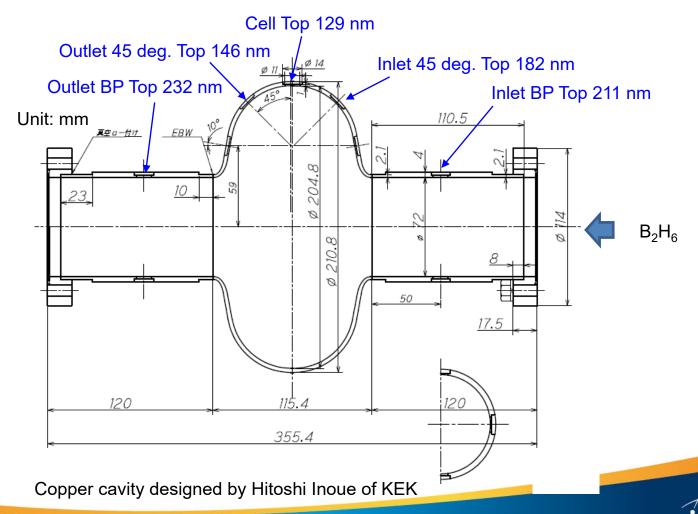


Recent result of thickness measurements of B films obtained in a test at TA-03-0040

- Used a spectroscopic ellipsometer alpha SETM
 - J.A. Woollam Co., Inc.
 - https://www.jawoollam.com/products/alpha-seellipsometer
 - Paolo Pizzol operated the ellipsometer.
 - The samples were from TA-35-213 Run 10 on 13-SEP-2013 (flowing B₂H₆ gas)
 - The zone 1 was at 300 C, but zones 2 and 3 were at 400 C. This probably reduced the thickness of B on the input BP.









Thanks for your attention!

I would like to thank Sakai-san, Okada-san, Kako-san and others at KEK for their help on this project!!

I hope we will be able to continue this project until we can produce MgB₂ cavities and test them at KEK.



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- Alp Findikoglu for measuring R_s of MgB₂ samples using parallel plate technique.



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- Jlab
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